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Spatial variations and temporal trends between 1994 and 2005 in polychlorinated biphenyls, organochlorine pesticides and heavy metals in European eel (*Anguilla anguilla* L.) in Flanders, Belgium

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Although tissue concentrations of PCBs, pesticides and heavy metals decreased over time, they remain sufficiently high for the consumption of wild captured eel to still be discouraged.

Abstract

In Flanders, the northern region of Belgium, European yellow eel muscle tissue was used as an indicator of environmental and potential human dietary exposure by hazardous chemicals of surface waters and sediments. Between 1994 and 2005, over 2800 eel captured at 365 stations were analysed for PCBs, pesticides and heavy metals. Contamination of eel in Flanders fell within the range of reported concentrations in other watersheds of Western Europe. A spatial analysis of the data demonstrated that the variation in pollutant concentration tended towards higher values. This was especially evident for PCBs, lindane, endrin, dieldrin and DDE. The concentration of almost all banned substances decreased significantly during the study period.

Keywords: *Anguilla anguilla*; Heavy metals; Organochlorine pesticides; Polychlorinated biphenyls; Linear mixed model

1. Introduction

The widespread occurrence of hazardous chemicals in the environment remains of major concern for wildlife and human health. Many chemicals, even banned ones, persist in the environment and continue to accumulate in ecosystems. In Europe, concern about the release of chemicals into the environment was shared by the European Parliament and the Council which adopted a far reaching Commission proposal aimed at ensuring greater safety in the manufacture and use of chemical substances (European Commission, 2006a). The new system REACH, which stands for registration, evaluation and authorisation of chemicals, will ensure that gaps in existing information on hazardous properties of chemicals are filled. In this renewed political context, the monitoring of chemicals in the environment and its ecosystems remains of crucial importance in order to produce data that serve as a baseline against which future policy results may be evaluated.

This paper presents a synthesis of a routine monitoring program that started in 1994 aimed at following the tissue concentration of hazardous chemicals in eel in Flanders, Belgium. Data were available for contamination of eel by polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) and heavy metals. PCBs and OCPs are persistent organic pollutants that are regulated under international agreements in order to reduce or eliminate their use and release into the environment. Heavy metals of concern include, amongst others, cadmium, lead and mercury because of their toxicity and their potential to bioaccumulate.

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The yellow eel was selected as a biomonitor for the aquatic environment for a number of reasons. Eel in the yellow stage are premature individuals. Eel do not reproduce in freshwater. Therefore, body burdens are not affected by a reproduction cycle and associated changes in lipid metabolism. Further, yellow eel have a high lipid content, increasing with age and reaching a maximum prior to silvering and emigration. They generally show life-long accumulation and low depuration rates (Larsson et al., 1991; Tulonen and Vuorinen, 1996; Knights, 1997; Daverat et al., 2006). Yellow eel are carnivores, widespread in all aquatic habitats, benthic, often burrowed in the bottom, tolerant to pollution and sedentary (Mason and Barak, 1990; Van Der Oost et al., 1996; Ashley et al., 2003; Linde et al., 2004). Home range may be larger in tidal estuaries than in freshwater habitats (Parker, 1995). Seasonal movements possibly occur while also the occurrence of erratic eel ('nomads') has been reported (Feunteun et al., 2003). These typical life history characteristics warrant the use of eel as an indicator for the presence of hazardous chemicals in the environment and, in particular, of those substances with a low solubility in water. The hypothesis is that the eel tissue concentration and body burden reflect well environmental exposure and that tissue concentrations are related to pollution levels of prey species, surface waters and sediments.

A second important advantage of the use of eel as bio-indicator of chemicals in the environment relates to the consequences of eel consumption for human health. Eel consumption is a definite pathway of human exposure to persistent organic chemicals and heavy metals (Harrad and Smith, 1999). Also in Flanders, explicit concern was raised in order to warn of the health hazard associated with the consumption of eel and other predatory fish species by recreational fishermen (Hoge Gezondheidsraad, 2005). Many of these chemicals are considered potential carcinogens and some are believed to disturb metabolic and endocrine functions of the human body (European Environmental Agency, 2005).

In addition, the European Commission (2006b) recently proposed a system to monitor a selection of priority substances and to report the chemical status of water bodies in order to protect aquatic life and human health. An important task was therefore to demonstrate the suitability of bio-indicators, such as the European eel (*Anguilla anguilla* L.), as models for evaluating the chemical status of surface waters which was required by the water framework directive. Eel contaminant profiles, especially for lipophylic substances, appeared to be a fingerprint of the contamination pressure of a specific site (Knights, 1997; Belpaire and Goemans, 2006).

So far, studies on the pollution of eel in Flanders focussed on targeted research actions (Roose et al., 2003; Versonnen et al., 2004; Goemans and Belpaire, 2004, 2005; Hoff et al., 2005; Maes et al., 2005; Belpaire and Goemans, 2006). A general synthesis reporting on all the data that were collected since 1994 has been provided as a report in Dutch (Goemans et al., 2003). This paper presents the first general description of the concentration of hazardous chemicals in the European eel in Flanders, the northern part of Belgium. Spatial information was provided at the river basin level while a selection of temporal data was retained in order to investigate temporal trends in the tissue concentration.

2. Material and methods

2.1. Field sampling and analysis of the lipid content

Between 1994 and 2005, 2839 eel were captured. Eel were always caught between March and October. Total annual catch varied between 25 eel in 1996 and 732 in 2000. On average, 237 eel were captured each year. Yellow eel were sampled at 365 different stations using fyke nets or an electrofisher. Stations were characterised as rivers or brooks, canals, polder water courses or closed water bodies such as old meanders, ponds or lakes. Stations were situated in all 11 river basins (Fig. 1). Between 1994 and 2005, 91 stations were visited twice for sampling; 16 stations were sampled three times, 6 stations were sampled four times and 2 stations were sampled five times. One station (Lake Weerde, a man-made water body) was sampled eight times between 1997 and 2005.

After capture, eel were sorted according to life history stage and only yellow eel were placed in cooling units for live transport to the laboratory. At the lab, eel were measured, weighed and stored at −20 °C for subsequent tissue analysis. From each individual eel six samples of muscle tissue (10 g wet weight each) were removed, labelled and frozen again at −20 °C. Two samples, originating from the mid part of the body, were analysed for heavy metals, OCPs and PCBs. Lipid was extracted and quantified using the Bligh and Dyer method (1959). The other samples were stored as back up.

2.2. Chemical analysis

2.2.1. Chemicals

Tissue samples were analysed for PCBs, several pesticides and nine heavy metals. Pesticides included two hexachlorocyclohexanes, three cyclodienes, hexachlorbenzene and three chloroethanes.

PCBs were used as insulating fluids in transformers, occur as plasticizers, oil and paint additives or as by-products of combustion. Tissue samples were analysed for 10 different congeners identified according to IUPAC numbers 28, 31, 52, 101, 105, 118, 138, 153, 156 and 180. Seven congeners are considered as indicator PCBs (28, 52, 101, 118, 138, 153, 180). In Belgium, the sum of these seven indicator PCBs (further abbreviated as $\sum$PCB) was used in national legislation to ensure food safety. The concentration of $\sum$PCB in fishery products may not exceed 75 ng g^{-1} wet weight. Hexachlorocyclohexanes were used as insecticides. Here, we report on α-HCH and γ-HCH (lindane), which were banned in Belgium in 2002. Cyclodienes in this study include dieldrin, endrin and chlordane. The use of dieldrin has been prohibited since 1974 while the use of endrin has never been authorised at all. Chlordane is a mixture of different components of which only transnonachlor was assessed. The use of the latter substance by agriculture has been prohibited since 1981 but non-agricultural use was allowed until 1998. Hexachlorobenzene (HCB) was formerly used as an fungicide and was banned in 1974. Concentrations of three chloroethanes (*p,p′*-DDD, *p,p′*-DDT, *p,p′*-DDE) were measured and their sum used as a proxy of total DDT ($\sum$DDT). DDTs have been banned since 1974 in case of agricultural application and since 1976 for all other uses. Finally, the levels of nine heavy metals (cadmium, lead, mercury, chromium, nickel, copper, zinc, arsenic, selenium) were determined.

2.2.2. Determination of PCBs and pesticides

Fish tissue was extracted using the Bligh and Dyer method (1959). The extract was evaporated (Rotavapor) and a minimum of 100 mg lipid was dissolved in hexane and applied on an aluminum oxide chromatography column. After elution with hexane, the lipid free eluate was evaporated and applied on a silica gel chromatography column. PCB congeners, *p,p′*-DDE and HCB were isolated after elution with hexane. After elution with diethylether/hexane (10/90) the remaining organochlorine pesticides were isolated. Both fractions were evaporated to 1 ml, after addition of an internal standard (tetrachloronaphtalene) and separated by gas chromatography using an

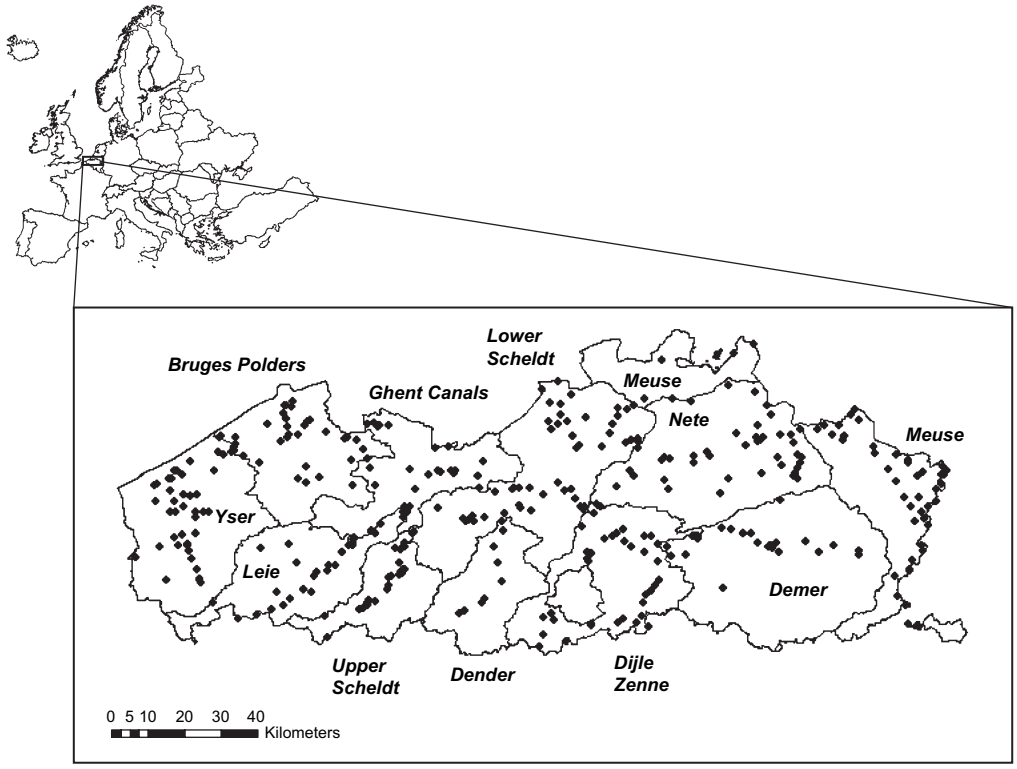

Fig. 1. Geographic position of Flanders in Europe and location of the sampling points of the eel pollutant monitoring network. The different sub-basins are indicated using their English names (if available).

Rtx-5ms capillary column (60 m × 0.25 mm × 0.25 μm), with helium as a carrier gas and an electron capture detector (ECD). The detection limit for both PCBs and pesticides was 0.5 ng g^{-1} lipid weight.

2.2.3. Determination of heavy metals

Fish muscle tissue (between 3 and 5 g) was placed in an oven for 12 h at 450 °C. Once cooled, 100 μl HNO_3 was added and the analyte was dried again at 450 °C for 1 h. Subsequently, 1 ml HNO_3 was added to the ash and diluted using distilled water.

Trace elements of Cr, Ni, Cu, Zn, Cd and Pb in solution were analysed using ICP-OES (Spectra AA-400 with Zeeman correction, Varian). The detection limits for each of these metals varied: 2 ng g^{-1} wet weight for Cd and Pb, 10 ng g^{-1} wet weight for Ni, 35 ng g^{-1} wet weight for Cr and 100 ng g^{-1} wet weight for Cu and Zn.

As and Se were determined using GF-AAS. Prior to analysis, fish tissue was heated in a medium of 5 ml HNO_3 and 3 ml H_2O_2 and afterwards diluted in distilled water. The detection limits for As and Se were 10 and 35 ng g^{-1} wet weight, respectively.

Hg was quantified using AAS (AMA 254 mercury analyser, Altec). Hg was detected if the concentration was higher than 10 ng g^{-1} wet weight.

2.2.4. Quality assurance

Analysis were carried out at two different Belgian research institutes (DVZ, the Sea Fisheries Department, Ostend and CODA, the Veterinary and Agrochemical Research Centre, Tervuren). Quality assurance consisted of the analysis of procedural blanks, reproducibility and repeatability tests, injection of standard solutions as unknowns, and analysis of certified reference material. Both institutes routinely analyse samples in the framework of the international proficiency testing scheme QUASIMEME (Quality Assurance of Information for Marine Environmental Monitoring in Europe) for organochlorines in biological samples and participate in intercalibration studies organised by the IAEA (International Atomic Energy Agency). Internal quality assurance at DVZ was realised by monthly analyses using certified reference materials. Gas chromatography equipment was calibrated every 60 samples and every 20 samples, two standards were analysed. Quality of heavy metal analyses performed at CODA was assured using reference materials and blanks every 12 samples (daily for Hg). ICP-OES and GF-AAS equipment was calibrated every 15 samples.

2.3. Data storage and statistical analysis

Eel contaminant data are stored in a database as a concentration in ng g^{-1} wet body weight. A unique identification number was assigned to each individual eel followed by a location code (linked to geographical information of the sampling station), sampling data (e.g. fishing date and procedure), length (cm), weight (g) and lipid content (as a percentage of wet weight). Concentrations of organic pollutants were imported on a lipid weight basis and converted into wet weight concentrations using lipid content as a conversion factor. Heavy metals were always expressed on a wet weight basis. Calculations used half the detection limit when a below detection limit reading was observed.

A general eel pollution profile was assessed using the average, the range and standard deviation based on all samples. Spatial analysis of sampling sites and the correlation structure between chemicals was investigated by arranging the data according to river basin. A linear mixed model was used to infer general trends in eel muscle tissue concentration over time and to test the null hypothesis that the slope of the trend line was not significantly different from zero. For this analysis, individual eel data based on wet weight were station-averaged and normalized using a $\log_{10}$ transformation. Only those stations that were sampled more than once were retained in the analysis. A paired *t*-test would be an appropriate statistic test if each station was sampled twice on two fixed dates. In this study, the data were, however, largely unbalanced with sets of two, three, four, five or eight repeated measurements, taken at different times and over different time intervals, hence the choice for a mixed model. The mixed model can be considered as a distinct linear regression for each set of clustered data with fixed and random, hence mixed, regression coefficients. In the model, time was considered as a fixed factor while the different stations constitute the random factor. The mixed model that was fitted through

the data was a random slopes and random intercepts model and has the following form:

$$\mathrm{Log}_{10}[C+1] = (\beta_0 + b_{0i}) + (\beta_1 + b_{1i}) \times (\mathrm{Year} - 1994) + \varepsilon_i \quad (1)$$

where β_0 and β_1 are regression parameters which were the same for all stations and b_0 and b_1 were station-specific regression coefficients; C was the concentration of a chemical substance on a wet weight basis. Essentially, we are only interested in the value for β_1 which describes the average trend over time. It was assumed that all random effects b_i are normally distributed with mean zero and variance $\sigma^2_{\mathrm{station}}$. The error term with residuals ε_i is normally distributed with mean zero and variance σ^2. $\sigma^2_{\mathrm{station}}$ is a 2×2 covariance matrix containing d_{11} the variance of the random intercepts, d_{22} the variance of the random slopes and $d_{12} = d_{21}$ which stands for the covariance between the random intercepts and the random slopes. Note that the covariance is related to the correlation between the random intercepts and the random slopes.

The MIXED procedure in SAS (SAS Institute Inc., 1999) was used to find parameter values for Eq. (1). The best solution was found using the restricted maximum likelihood estimator. It follows that mixed models are not interpreted in terms of explained variance (R^2). Inference for the parameters β_0 and β_1 was based on the Wald statistic. In the model, it was assumed that the covariance matrix was unstructured by entering the SAS statement "type = un" in the code. Verbeke and Molenberghs (2000) give statistical details and examples.

3. Results

3.1. Length, weight and bioaccumulation

A total of 2839 eel was analysed for at least one of the substances listed in Table 1. Mean length and weight of eel included in this analysis were 41.8 ± 9.3 cm and 153.5 ± 152.7 g, respectively. Lipid content of the total body weight averaged $14.9 \pm 10.2\%$. Eel tissue concentrations (mean, range, standard deviation) of PCBs, OCPs and heavy metals are presented in Table 1.

PCBs were ubiquitous in eel. The average concentration of the sum of the seven indicator PCBs was 605 ng g^{-1} wet weight, exceeding the Belgian limit for human consumption (75 ng g^{-1} wet weight) almost by one order of magnitude (Table 1). Note the high standard deviation and the maximum concentration of almost 12 500 ng g^{-1} wet weight. The distribution of $\sum$PCB in eel was positively skewed, so the probability of capturing highly contaminated eel was higher than that could be expected in a normal distribution. Seventeen per cent of all individuals displayed a total PCB concentration of >1000 ng g^{-1} wet weight and 1.7% with concentration higher than 5000 ng g^{-1} wet weight. Note that the highest levels were found for PCBs 138, 153 and 180 which are particularly recalcitrant compounds (Knights, 1997).

Concentrations of the biocide lindane ranged between 0.01 and 2225 ng g^{-1} wet weight with an average concentration of 27.9 ng g^{-1} wet weight. Concentrations of α-HCH averaged 0.64 ng g^{-1} wet weight. Dieldrin and endrin, once used as insecticides but banned since 1974, were detected in >90% of all eel analysed. Values of dieldrin averaged 15.6 ng g^{-1} wet weight with a maximum of 389 ng g^{-1} wet weight; mean endrin concentration was 1.4 ng g^{-1} wet weight. The fungicide hexachlorobenzene was detected in all eel at an average concentration of 5.9 ng g^{-1} wet weight (max 192 ng g^{-1} wet weight). DDTs were present in all fish with $\sum$DDT varying between 1.5 and almost 4000 ng g^{-1} wet weight. The distributions of pesticide concentrations were also positively skewed.

Eel carried significant concentrations of heavy metals in their muscle tissue. Concentrations of mercury, cadmium and lead, substances for which maximum limits apply on a European level, averaged 116, 15.8 and 81 ng g^{-1} wet weight, respectively. Maximum observed concentrations for each of these metals exceeded or were well over the European maximum residue limit (MRL). Eel also exhibited extreme values for other heavy metals (Table 1), exceeding the average concentration by an order of magnitude, demonstrating the high potential of eel for severe contamination.

Table 1
Mean eel life history statistics and mean muscle tissue concentration and range (ng g^{-1} wet weight) of different pollutants in muscle tissue sampled in surface waters of Flanders (Belgium)

	N	Mean	Minimum	Maximum	St. Dev.
Eel life history data					
Length (cm)	2839	41.79	19.2	102.30	9.28
Weight (g)	2838	153.46	11.7	2284.00	152.69
Lipid content (%)	2528	14.92	0.52	57.59	10.18
Substance					
PCBs					
PCB 28	2525	6.44	0.0035	292.65	15.11
PCB 31	2525	3.04	0.0037	211.84	7.97
PCB 52	2526	30.61	0.0087	624.36	53.82
PCB 101	2526	55.55	0.0272	1505.79	104.77
PCB 105	2526	18.12	0.0104	478.12	34.81
PCB 118	2526	57.13	0.2904	2076.45	112.23
PCB 138	2526	149.69	0.5805	2924.25	295.65
PCB 153	2526	211.89	1.0423	5098.68	430.33
PCB 156	2525	13.98	0.0263	352.71	25.17
PCB 180	2525	93.48	0.1250	2131.50	180.93
$\sum$PCB	2524	604.99	3.5213	12455.38	1118.56
Pesticides					
α-HCH	2528	0.64	0.1	16.94	1.32
Lindane	2527	27.89	0.0109	2225.46	131.68
Dieldrin	2528	15.63	0.0046	388.78	30.21
Endrin	2446	1.39	0.0026	495.83	11.59
HCB	2526	5.89	0.0026	191.95	8.91
TDE	2528	26.26	0.0108	568.46	41.36
p,p′-DDT	2528	3.19	0.0037	187.81	9.58
p,p′-DDE	2526	61.77	0.1007	3422.63	112.73
$\sum$DDT	2528	90.77	1.5149	3995.42	148.27
Transnonachlor	2528	1.43	0.0026	52.03	2.73
Aldrin	548	1.11	0.0056	14.11	2.21
Heavy metals					
Mercury	2769	116.62	5.0	1185	98.89
Cadmium	2809	15.75	1.0	2474	62.21
Lead	2802	81.17	1.0	3453	172.17
Copper	2117	909.73	50.0	436,000	10,006.90
Zinc	2117	25,864.79	1200.0	243,100	15,919.30
Nickel	2117	207.83	5.0	16,300	692.00
Chromium	2117	254.51	17.5	13,690	455.97
Arsenic	1410	168.13	14.0	1805	176.72
Selenium	1410	753.93	25.0	5098	499.75

Number of eel (*N*), mean, range (minimum–maximum) and standard deviation (St. Dev.) were calculated for the period 1994–2005.

3.2. Spatial variability in the tissue concentration

Figs. 2–4 summarize the spatially resolved pollutant averages on a river basin level (Fig. 1). Two major findings emerged: some pollutants were evenly spread while others clearly peaked in selected river basins and the variance around the mean was high due to the presence of high concentrations in some specific sites. The contamination of eel by different pesticides was most notable in the basin of river Yser. This was particularly evident for lindane with a basin average of 262.9 ± 552.7 ng g^{-1} wet weight. Also α-HCH and dieldrin peaked in this river basin with averages that were clearly higher than the overall average tissue concentration (Table 1). PCB contamination more or less increased along a west–east gradient and reached a maximum in the basin of river Meuse where an average ∑PCB of nearly 800 ng g^{-1} wet weight was detected. DDT in eel muscle tissue peaked in the Upper Scheldt and Demer river basins, while Cd and Pb pollution was typical for the River Nete. Pollution levels of As and Hg, as well as other pesticides such as HCB and chlordane, were more evenly spread throughout the entire region.

3.3. Temporal trends in the tissue concentration

Sampling took place more than once at 116 stations, and these data were useful to investigate time trends in eel pollutant concentration. Sampling station-averaged tissue concentration data in a general linear mixed model was used to infer an average time trend of pollutant contamination. Station-averaged time profiles and the fitted regression line for several organochlorine pollutants and four metals between 1994 and 2005 are presented in Figs. 5–7. Model parameters (intercept and slope) and diagnostics are given in Table 2. There were significant reductions in the average wet weight concentration of all PCB congeners, nearly all pesticides and four metals. In Table 2, the variance present in the data set was partitioned into the variance due to the random intercepts, variance due to the random slopes, covariance between these parameters

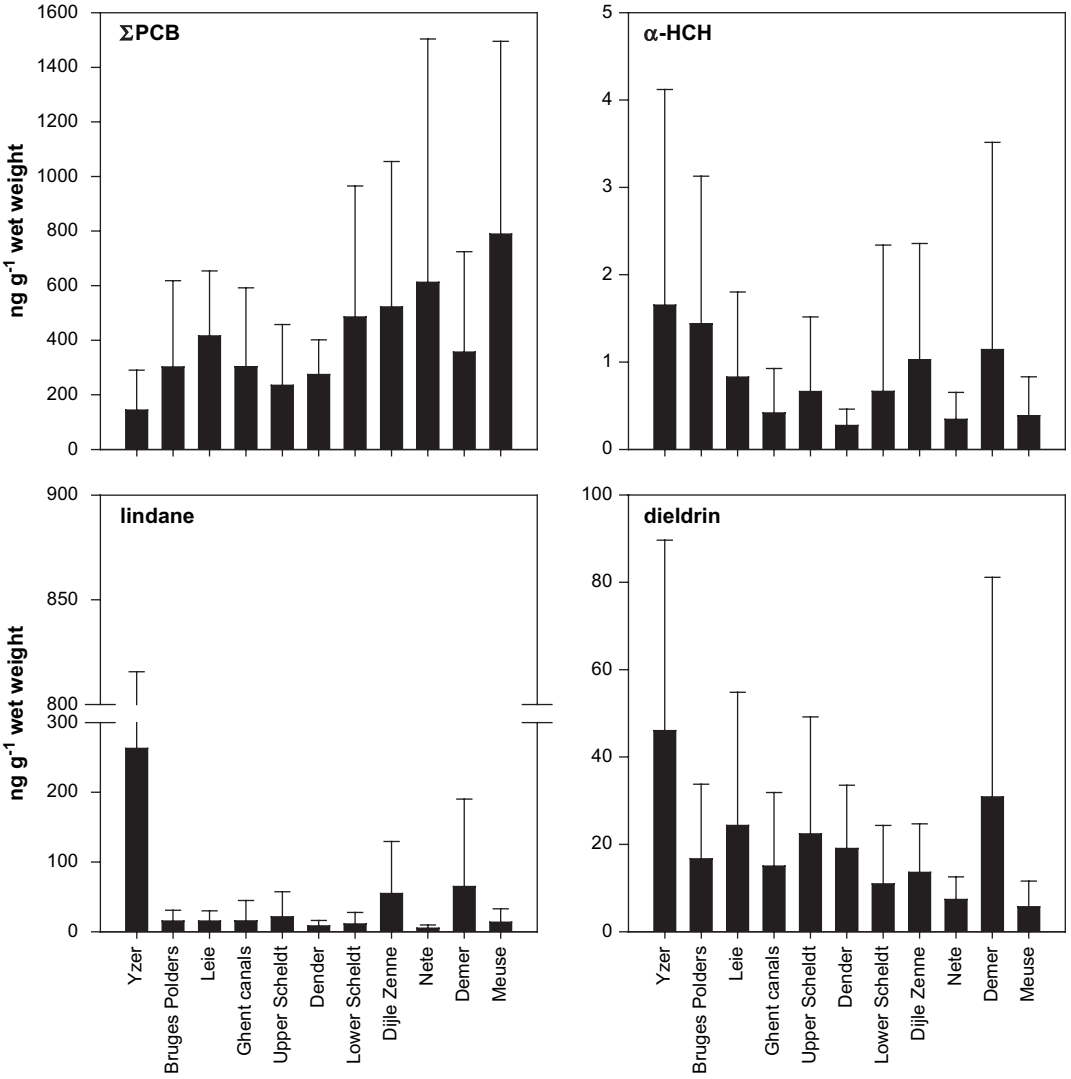

Fig. 2. Spatial distribution of the average eel muscle tissue concentration of ∑PCB, lindane, α-HCH and dieldrin over the different river basins in Flanders (Belgium).

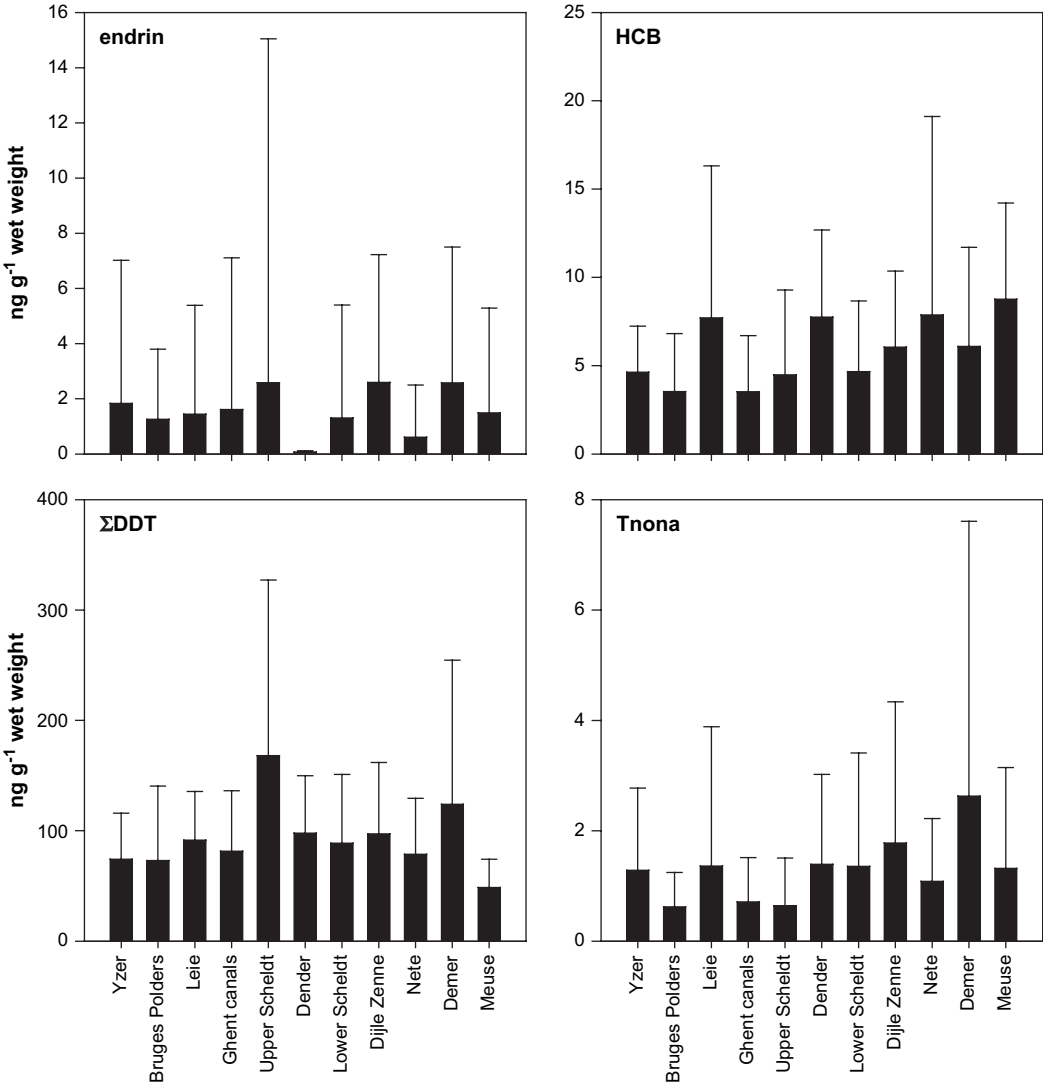

Fig. 3. Spatial distribution of the average eel muscle tissue concentration of endrin, ∑DDT, HCB and transnonachlor (Tnona) over the different river basins in Flanders (Belgium).

and, finally, the variance of the residuals. Generally, most of the variance was due to the random intercepts, which corresponds to variability in tissue concentration amongst the different sampling stations. Further, the covariance between intercepts and slopes was invariantly negative. This means that in stations where pollutant concentrations were initially above average, the rate of reduction was more pronounced than in stations with initially below-average concentrations.

All PCB congeners had significantly negative slopes indicating their gradual reduction in the freshwater environment. Fig. 5 presents the station-averaged time profiles as well as the modelled trend line for PCB 153 and ∑PCB. Based on the regression model, the back calculated average ∑PCB concentration of eel was 770.6 ng g^{-1} wet weight in 1994 while for 2005, the regression model predicted an average concentration of 125.3 ng g^{-1} wet weight. It follows that the PCB concentration of eel decreased with a modelled rate of 15% per year.

Also concentrations of most pesticides decreased significantly over time. This was especially evident for α-HCH and lindane (Fig. 6). Similar reductions were modelled for HCB, dieldrin and endrin. Unexpectedly, concentrations of *p*,*p*′-DDT increased over time but this effect was countered by significant reductions of the metabolites *p*,*p*′-DDE and *p*,*p*′-DDD (=TDE). As a result, ∑DDT also decreased significantly.

The most notable reductions in the tissue concentration of heavy metals were observed for lead (Fig. 7), arsenic, nickel and chromium. However, for the latter three metals, the trend may be biased as data were available only since 2000. The selenium concentration increased significantly but, similarly, data were available only since 2000. No trend was observed in the concentrations of mercury and cadmium (Fig. 7).

4. Discussion

Belgium, and in particular its northern region Flanders, suffers substantial environmental problems. As a result, the country performs poorly in international studies. Based on

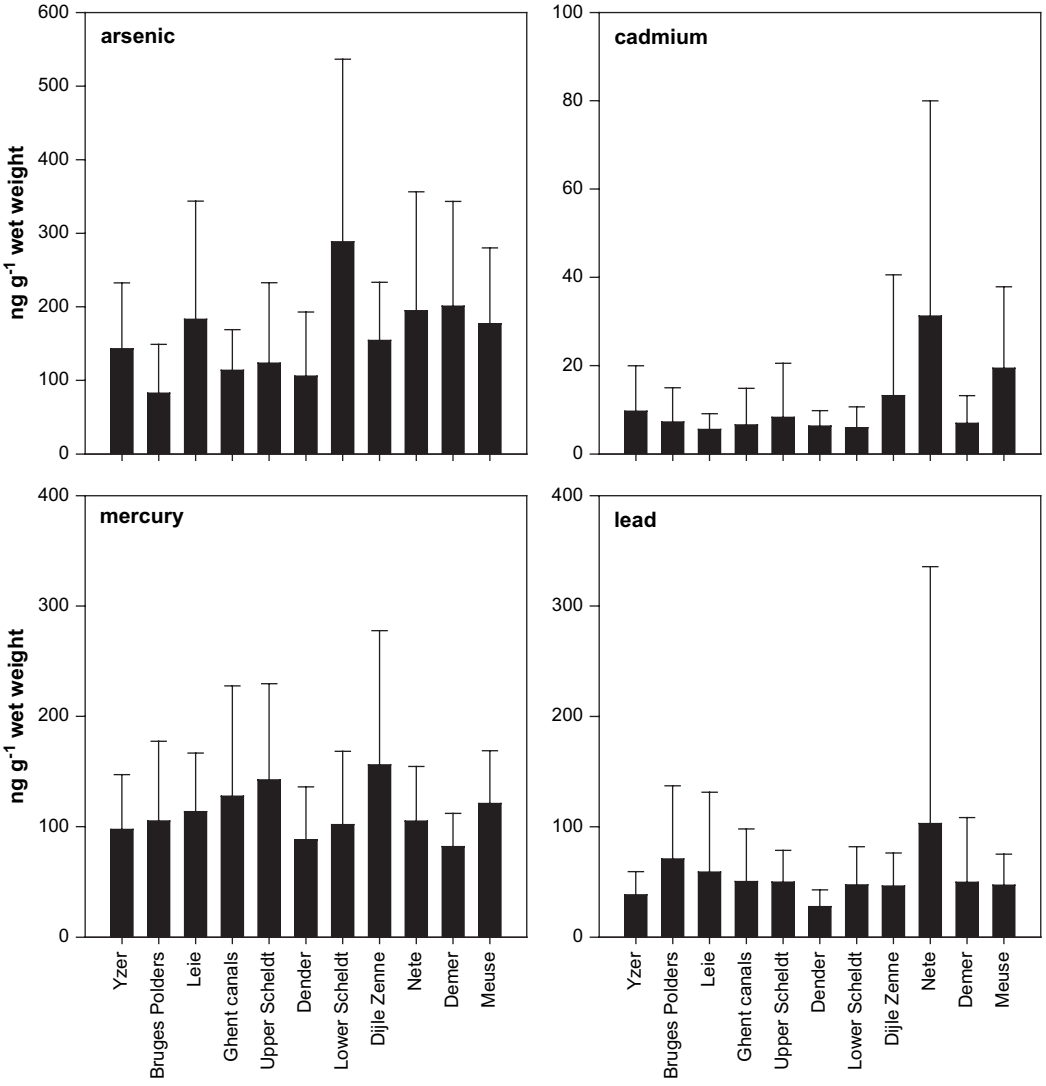

Fig. 4. Spatial distribution of the average eel muscle tissue concentration of four heavy metals over the different river basins in Flanders (Belgium).

a recently established environmental performance index (Esty et al., 2006), Belgium ranks 39th globally, last within the EU-25 and 26th out of 29 OECD member countries. Essentially, environmental problems in Flanders relate to the high human population density coupled with a historical lack of proper land use planning. The landscape is characterised by a patchy distribution of urbanization, industry, agriculture and nature. The high population density, as well as intensive livestock production, and the economical development of chemical industries result in a high pressure on the environment while at the same time, the fragmentation of the landscape greatly reduces the possibilities of targeted actions and effective environmental management. In particular, the management of water resources is an issue of concern. Belgium lags behind in the EU and faces serious water challenges. An additional problem is the trans-boundary aquatic pollution from neighbouring countries via the main rivers (Scheldt and Meuse).

This paper compares the pollution profile of European eel caught in Flemish inland waters with profiles reported elsewhere in Europe, using peer-reviewed papers that presented quantitative contaminant data in eel muscle tissue. Where appropriate, results were averaged while concentrations below detection limits were not considered. The results of this literature survey, as well as references, were listed in Tables 3–5. This comparison is not complete so we refer to Bruslé (1990) for additional concentration ranges of heavy metals and to Robinet and Feunteun (2002) for mean concentrations and ranges of different synthetic chemicals in yellow European eel muscle tissue. Further, the comparison between our results and published data was likely biased. Almost all the references that are included in Tables 3–5 focus on particular sites, where contamination was suspected, or are limited to single river. In addition, reported concentrations are based on sample sizes that were considerably smaller than those in this study. Arguably, it was appropriate to also include our basin-specific and individual maximum values in this comparison.

Average PCB contamination levels of Flemish eel fall within concentrations reported for Western Europe. Data

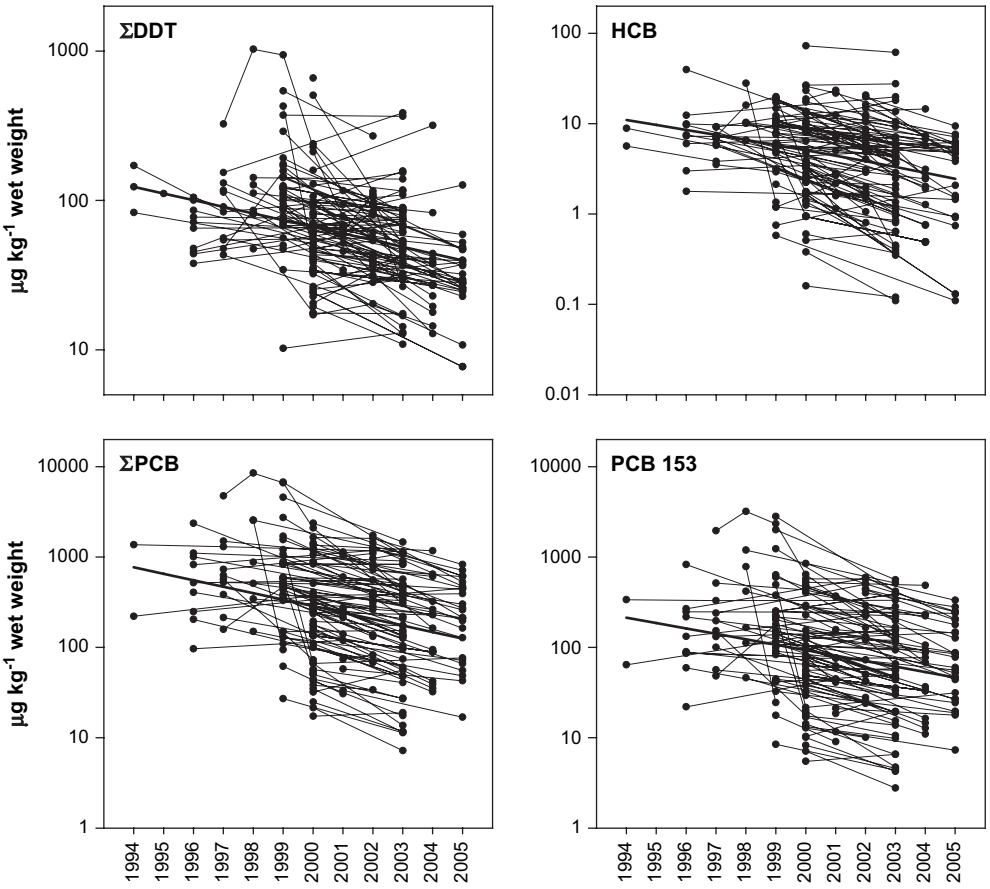

Fig. 5. Temporal trends in average eel muscle tissue concentration of ∑DDT, ∑PCB, HCB and PCB 153 at sampling stations that were sampled more than once between 1994 and 2005. The bold line represents the average time trend which was modelled using a linear mixed model. See Table 2 for the intercept and slope.

obtained from literature refer to inland waters of Spain, Luxembourg, UK and The Netherlands. Average PCB concentrations in this study were higher than those reported for river Turia (Spain), the Severn (UK) and the Sur (Luxembourg) but comparable to those of eel caught in the waters nearby Amsterdam (The Netherlands). Eel caught in river Moselle (Luxembourg) indicated heavy PCB contamination. Individual maximum levels of the different PCB congeners that were found in this study were, however, at least one order of magnitude higher than the average concentrations given in Table 3. The proportion of samples analysed in this study that exceeded the maximum reported average concentration based on the literature reports varied between 1.1% for PCB 52 and 10.4% for PCB 156 (Table 3). This demonstrates that eel experienced substantial exposure during the study period. In particular, eel captured in the basin of River Meuse were highly loaded with PCBs. River Meuse runs through an important industrial area including energy production and power transformation industries, which are possible historical sources of PCB contamination. The PCB data reported for the Netherlands by De Boer and Hagel (1994) proved to be particularly relevant to this analysis given the similarities in sampling design. Our study differs with De Boer and Hagel (1994) in that the Dutch monitoring program mainly focussed on contaminated sites in the rivers Rhine and Meuse, sampled between 1977 and 1990. Our data were collected 15 years later and also included smaller sized rivers and brooks, where exposure was assumed to be lower. Yet, we argue that the data presented in De Boer and Hagel (1994) may be used as a baseline against which the present PCB pollution in Flemish eel can be evaluated. Average values reported in this study for individual PCB congeners were four times lower than the average results presented in De Boer and Hagel (1994). Then again, it appeared that 7.7% of our tissue samples tested for PCB 153 exceeded the baseline value based on De Boer and Hagel (1994) (Table 3). For PCB 156 and PCB 180, the proportion of eel with total concentrations above the average concentrations reported by De Boer and Hagel (1994) was 10.4% and 13%, respectively. So, in spite of decreasing concentrations, still a significant proportion of eel that was present in Flemish inland waters during the study period had a relatively high PCB concentration.

Reported contaminant concentrations in European eel by different pesticides were variable (Robinet and Feunteun, 2002). Apart from endrin, average concentrations in the present study did not exceed any of the means reported for other surface waters in Europe (Table 4). However, pesticide contamination in eel muscle tissue was not evenly distributed in Flanders with high levels of lindane and dieldrin in the basin

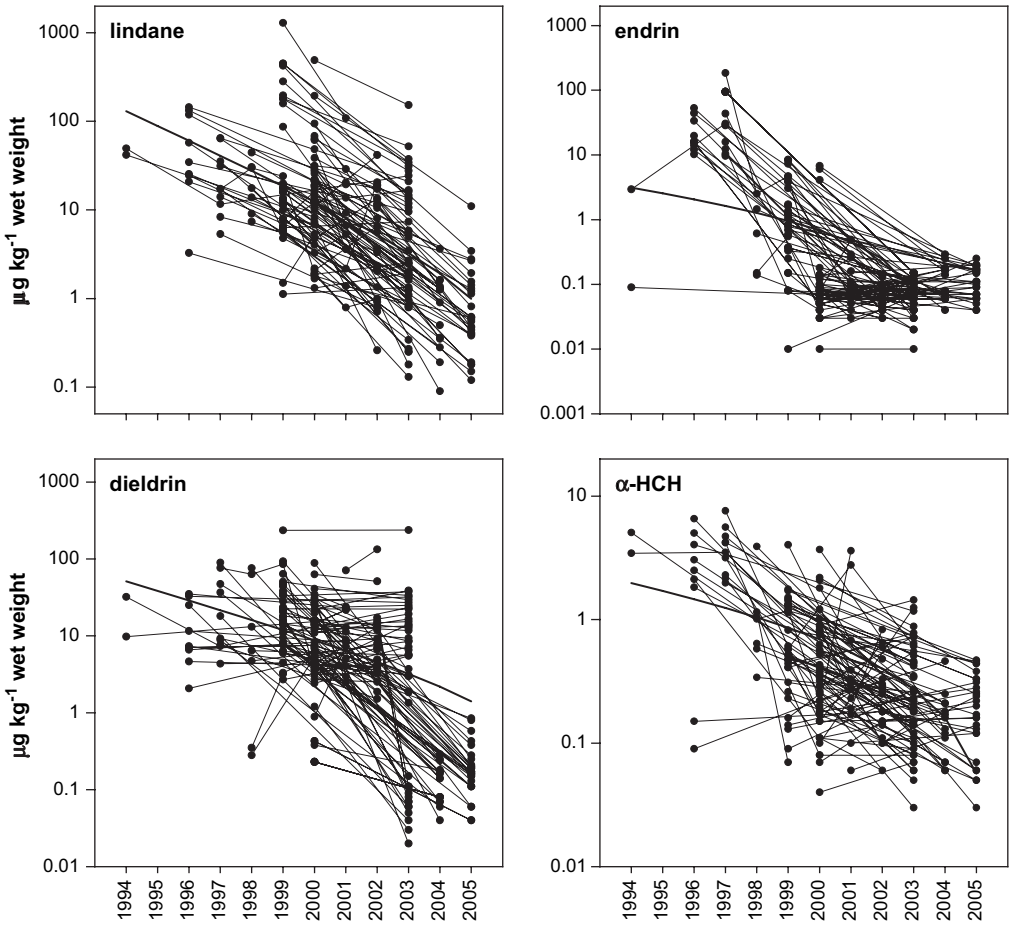

Fig. 6. Temporal trends in average eel muscle tissue concentration of lindane, dieldrin, endrin and α-HCH at sampling stations that were sampled more than once between 1994 and 2005. The bold line represents the average time trend which was modelled using a linear mixed model. See Table 2 for the intercept and slope.

of River Yser and above average DDT concentrations in the basin of the Upper Scheldt. Using these basin-averaged data (Figs. 2–3), it appeared that pesticide tissue concentrations reported in this study are at the higher end relative to values reported for the rest of Europe. Most data in Table 4 refer to results for DDT derivatives and lindane. The incidence of the latter pesticide was highest in the Severn and in the delta of river Rhone. In Flanders, lindane and dieldrin peaked in the basin of River Yser, where land use is predominantly agriculture. Ten sampling sites showed tissue concentrations >100 ng g^{-1} wet weight, evidencing high local body burden in fish. *p,p′*-DDE varied in Europe between 3.9 and 187.9 ng g^{-1} wet weight and averaged 56 ng g^{-1} wet weight which was considerably lower than the average that was observed in the basin of the Upper Scheldt.

Only limited information of contamination by heavy metals in eel tissue was available in the literature. Bruslé (1990) reviewed metal contamination ranges in eel. Again, average concentrations do not differ much from the reported ones as the pollution was very much focussed in particular river basins. This was especially evident for Cd and Pb, which peaked in the basin of River Nete. The pollution in this river basin can be related to the presence of different non-ferrous industries producing zinc, cadmium and copper. As a consequence, heavy metals have been widespread in the local environment.

In summary, average contamination of eel in Flanders falls within the range of reported concentrations in other watersheds of Western Europe. However, spatial partitioning of the data demonstrated that the variation in pollutant concentration was positively skewed. This was especially evident for PCBs, lindane, endrin, dieldrin and DDE. A similar conclusion was made for heavy metals.

4.1. Trends in eel contamination

In this paper, evidence was presented that the tissue concentration of some persistent chemicals in European eel has declined. Time series of the tissue concentration of PCBs and several pesticides showed a negative time trend. This conclusion was based on the application of linear mixed models for longitudinal data. This method was preferable to regressing annual averaged concentrations over time since the data were clustered according to sampling stations and hence, not independent from each other. The analysis demonstrated that river basins and sampling sites had clearly different pollution profiles.

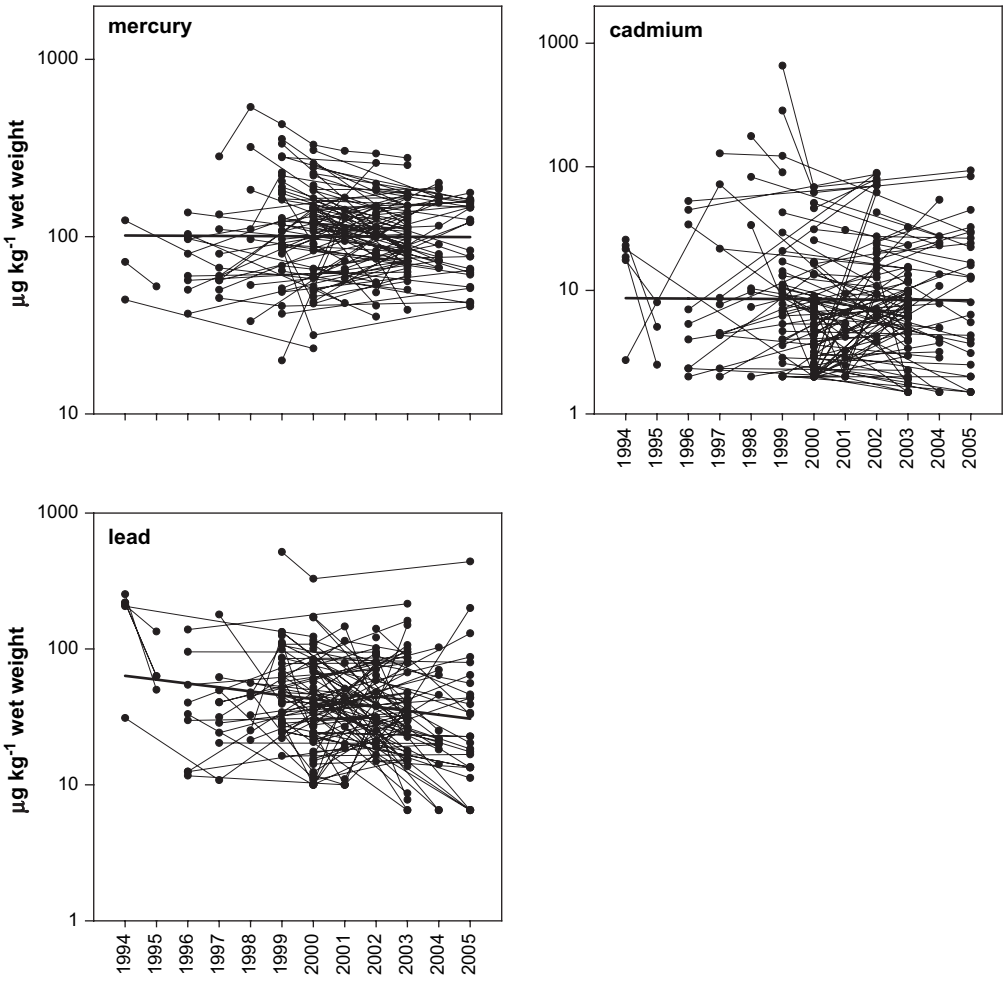

Fig. 7. Temporal trends in average eel muscle tissue concentration of three heavy metals at sampling stations that were sampled more than once between 1994 and 2005. The bold line represents the average time trend which was modelled using a linear mixed model. See Table 2 for the intercept and slope.

The observed decline of PCBs in eel tissue was in agreement with other studies reporting on time series of contaminants in fish. PCBs were banned from the EU in 1985 and since then, several time series have indicated decreasing levels of contamination. A well-known example was the decreasing trend of PCBs in human breast milk in Sweden (Noren and Meironyte, 2000). In teleosts, significant declines are reported for Spanish commercial fishes between 1995 and 2003 (Gomara et al., 2005), in salmonids of Lake Michigan between 1972 and 1994 (Lamon et al., 1998) and in Arctic char for the period 1960–1996 in Lake Vattern, Sweden (Lindell et al., 2001). In Flanders, concentrations of $\sum$PCB in eel tissue were shown to have decreased by 15% per year. This rate was in agreement with other studies in fish (Lindell et al., 2001). Also the time series of lindane, α-HCH, dieldrin, endrin, and HCB showed that bans and environmental policies lead to decreased concentrations.

A notable exception to this general decrease in persistent organic pollutants was *p*,*p*′-DDT. The linear model indicated an increase while at the same time, *p*,*p*′-DDD and *p*,*p*′-DDE showed significant decreases. However, it appeared that *p*,*p*′-DDT decreased between 1994 and 2001 while concentrations increased again after 2002. At first sight, the ratio between DDE and DDT was in all eel analysed >1, suggesting that remaining DDT had not been recently reapplied. However, at some locations in Flanders (Kanaal Dessel Schoten, Handzamevaart and Ieperkanaal) the ratio between DDE and DDT rapidly decreased over a few years by an order of magnitude of three. Such a steep decrease, even if the ratio was higher than one, probably indicates recent application of DDT and shows that not all stock was depleted. These results, as well as the recent observation of the human blood samples, particularly of the juvenile population living outside urban areas, still containing DDT (Steunpunt Milieu en Gezondheid, 2006) should urge regional policy makers to make a serious attempt in order to collect remaining stock of banned pesticides.

Mercury, cadmium and lead are heavy metals of special concern as they tend to bioaccumulate in the body. This study showed that only the concentration of lead in eel muscle tissue was consistently decreasing between 1994 and 2005, which

Table 2
Linear mixed models results and diagnostics of the regressions of the concentration of contaminants and pesticides based on wet weight against time

	N	β_0	β_1	t	P	d_{11}	d_{12}	d_{22}	σ
PCBs									
PCB 28	256	0.8376	−0.04317	−5.21	<0.0001	0.3195	−0.02793	0.003641	0.03427
PCB 31	256	0.6425	−0.03547	−5.18	<0.0001	0.1921	−0.01783	0.002404	0.02465
PCB 52	256	1.6996	−0.08075	−11.29	<0.0001	0.2484	−0.00309	0.001051	0.04559
PCB 101	256	1.9204	−0.07721	−10.61	<0.0001	0.2519	−0.00428	0.000721	0.05274
PCB 105	256	1.4554	−0.06607	−11.84	<0.0001	0.1775	−0.00559	0.000584	0.02855
PCB 118	256	1.9132	−0.0696	−11.22	<0.0001	0.2062	−0.00521	0.000744	0.03487
PCB 138	256	2.2845	−0.07387	−10.11	<0.0001	0.3354	−0.00876	0.000652	0.05414
PCB 153	256	2.3323	−0.05965	−8.03	<0.0001	0.3275	−0.0077	0.000726	0.05494
PCB 156	256	1.1426	−0.04289	−7.51	<0.0001	0.1887	−0.00342	0.000319	0.03424
PCB 180	256	1.9934	−0.06046	−8.18	<0.0001	0.316	−0.0056	0.000505	0.05767
∑PCB	256	2.8874	−0.07143	−10.13	<0.0001	0.2781	−0.00333	0.000549	0.05103
Pesticides									
a-HCH	256	0.4736	−0.04087	−11.06	<0.0001	0.05387	−0.00471	0.000318	0.01793
Lindane	256	2.118	−0.1653	−18.18	<0.0001	0.4585	−0.02463	0.000964	0.09491
Dieldrin	256	1.7184	−0.1215	−10.4	<0.0001	0.2883	−0.03071	0.004721	0.1112
Endrin	249	0.6146	−0.06479	−7.04	<0.0001	0.4175	−0.04744	0.005242	0.05075
HCB	256	1.0777	−0.04906	−8.67	<0.0001	0.1294	−0.0041	0.000435	0.03263
TDE	256	1.4564	−0.03919	−3.14	0.0022	0.4285	−0.05093	0.007281	0.1009
p,p′-DDT	256	−0.4265	0.1082	9.74	<0.0001	0.2697	−0.04518	0.007798	0.0659
p,p′-DDE	256	2.0327	−0.05871	−9.85	<0.0001	0.1329	−0.00755	0.000672	0.03453
∑DDT	256	2.0939	−0.04403	−7.19	<0.0001	0.1041	−0.00442	0.000384	0.04192
Transnonachlor	256	−0.2373	0.07239	8.64	<0.0001	0.1892	−0.02658	0.004179	0.0356
Heavy metals									
Mercury	266	2.0107	−0.0008	−0.16	0.8708	0.08724	−0.00648	0.000637	0.02176
Cadmium	268	0.9845	−0.00143	−0.17	0.8645	0.1502	−0.00323	<0.0001	0.09567
Lead	268	1.8097	−0.02813	−3.16	0.0021	0.09827	−0.00915	0.001597	0.09254
Copper	191	2.6312	0.009044	1.2	0.2351	0.07991	−0.00456	<0.0001	0.03505
Zinc	191	4.4179	−0.00777	−1.29	0.2033	<0.0001	−0.00289	0.000823	0.01489
Nickel	191	2.4514	−0.08422	−4.55	<0.0001	0.6791	−0.0376	<0.0001	0.21715
Chromium	191	2.5072	−0.02356	−2.32	0.0237	0.02888	−0.00125	<0.0001	0.05499
Arsenic	160	2.5802	−0.05309	−3.95	0.0003	0.03658	−0.00548	<0.0001	0.02548
Selenium	160	2.3095	0.05859	6.21	<0.0001	0.1111	−0.00418	<0.0001	0.02592

The table shows for each pollutant the total number of samples, the model intercept β_0 and slope β_1, the t-value and significance level P corresponding to the slope and the partitioning of the variance over d_{11} (variance of the random intercepts), d_{22} (variance of the random slopes), d_{12} (covariance between random intercepts and random slopes) and σ (residual variance). Scatterplots are presented in Figs. 5–7. No model was constructed for aldrin due to limited data.

possibly related to the gradual changeover from leaded to unleaded fuels and a reduction of industrial emissions. Cadmium and mercury, however, remain common environmental pollutants in the industrialized region of Belgium as there was no evidence that exposure of eel to these metals was decreasing. For other metals, data were available only since 2000 so a continuation of the sampling program is necessary to confirm the observed trends.

4.2. Eel as pathway of human exposure to pollutants

Both European and national legislative initiatives have established a framework on maximum residue and contaminant levels in or on food and feed of plant and animal origin including the Regulation (EC) No. 396/2005 of the European Parliament and of the Council (European Commission, 2005). The maximum pesticide residue level (MRL) in foodstuffs is 0.01 mg kg^{-1}. This general limit is applicable by default, i.e. in all cases where an MRL has not been specifically set for a product or product type. Definitive tolerances will be listed in Annex II of the regulation which is yet to be published. Until then, MRLs for pesticides in products of animal origin established by Council Directive 86/363/EEC, as amended, are in force (European Commission, 1986). For cadmium, lead and mercury, levels have been established by Commission regulation (EC) No. 466/2001 (European Commission, 2001). In February 2006, the European Commission (2006c) revised the maximum levels for dioxins and dioxin-like PCBs in foodstuffs (Commission regulation (EC) No. 199/2006). These limits evidently apply in Belgium, a member state of the EU. However, the maximum limit for PCBs differs in Belgium in that the sum of the seven indicator PCBs was used to adopt a maximum level. The European legislative framework was used in this paper in order to assess the consumption quality of eel (Table 6). In particular, the proportion of eel analysed in this study that exceeded maximum residue or contaminant limits was calculated.

Relative to maximum quantities as adopted by legislation, it appears that eel tissue was, in general terms, compliant

Table 3
Reported concentrations of PCBs in *Anguilla anguilla* in Europe

Sampling site	Year	28	31	52	101	105	118	138	153	156	180	Reference
Amsterdam area (The Netherlands)	1991	17.63		28.53	38.49		74.93	115.37	112.90		38.65	Van Der Oost et al. (1996)
Inland waters (The Netherlands)	1977–1990	28.71	12.33	261.44	354.90	49.08	226.17		570.93	29.56	181.98	De Boer and Hagel (1994)[a]
River Moselle (Luxemburg)	1996–1997						179.40	694.60	783.90		335.70	Dauberschmidt and Hoffmann (2001)
River Severn (UK)	1996			8.65	6.30	3.75	12.65	26.60	28.15	1.95	12.05	Harrad and Smith (1999)
River Sur (Luxemburg)	1996–1997						5.70	29.00	39.40		16.80	Dauberschmidt and Hoffmann (2001)
River Turia (Spain)	2000	0.75		2.16	2.99		2.74	12.30		0.35	2.43	Bordajandi et al. (2003)
Proportion of eel in Flanders > Max	1994–2005	6.2%	6.1%	1.1%	1.9%	6.9%	3.8%	3.6%	4.9%	10.4%	5.5%	

Numbers refer to different congeners. The last row presents the proportion of eel (%) captured in this study of which tissue concentrations exceeded the maximum value reported in the cited studies (Max).
[a] Value represents average of the data.

with European regulations for pesticides and heavy metals. The incidence of different pesticides in fish tissue was related to land use, so concentrations of lindane and dieldrin peaked in the western part of the country (the basin of river Yser) which has intensive horticulture. In that basin 14% of eel are non-compliant for lindane.

PCB concentrations in eel muscle tissue remain problematic. About 76% of the analysed individuals and 78% of the sampling stations exceeded the maximum level for human consumption. This maximum was based on the sum of seven indicator PCBs which, in Belgium, was fixed at 75 ng g^{-1} wet weight basis for fish. This limit was established after the Belgian dioxin crisis in 1999. In the spring of 1999, dioxin was introduced into the Belgian food chain via contaminated animal fat that was used in animal feeds. Due to the subsequent awareness of the public to food safety issues, the consumption of eel caught in public waters by anglers was prohibited. However, this ban was lifted again in December 2005. In February 2006, the European Commission established a maximum level for the sum of dioxins and furans in muscle meat of eel (4.0 pg WHO-PCDD/F-TEQ g^{-1} wet weight) as well as a maximum for the sum of dioxins, furans and dioxin-like PCBs (12.0 pg WHO-PCDD/F-PCB-TEQ g^{-1} wet weight) (European Commission, 2006c). Only two PCB congeners that were included in this study have toxic equivalent factors for human risk assessment. Neither the most toxic dioxin-like PCBs (congeners 126 and 169) nor dioxins and furans were monitored in this study. Therefore, it was not possible to directly assess the potential risks of consuming eel. As an alternative,

Table 4
Reported concentrations of organochlorine pesticides in *Anguilla anguilla* in Europe

Sampling site	Year	α-HCH	Lindane	Dieldrin	Endrin	HCB	TDE	*p,p′*-DDT	*p,p′*-DDE	t-DDT	Tnona	Reference
Amsterdam area (The Netherlands)	1991	39.1	1.3	0.6	0.4	59.1	0.6	47.3	97.7			Van Der Oost et al. (1996)[a]
Ebro delta (Spain)	1985						48.8	15.8	35.9			Ruiz and Llorente (1991)
River Moselle (Luxemburg)	1996–1997		37.4						187.9			Dauberschmidt and Hoffmann (2001)
Orbetello lagoon (Italy)	2002		10.0			0.1			3.9	4.4		Corsi et al. (2005)[a]
Po delta (Italy)	1994					0.2	19.9	4.2	27.4			Bressa et al. (1997)[a]
River Severn (UK)	1996		2210.1									Harrad and Smith (1999)[a]
River Sur (Luxemburg)	1996–1997		53.1						42.8			Dauberschmidt and Hoffmann (2001)
River Turia (Spain)	2000						8.5	7.0	29.9	45.3		Bordajandi et al. (2003)[a]
Vaccares lagoon (France)	1996–1997		120.0	0.6		5.6			6.1	107.6		Roche et al. (2000)[a,b]
Vaccares lagoon (France)	1998		266.7	54.7								Roche et al. (2002)[a,b]
River Vanajavesi (Finland)	1990–1993	2.5	4.8			3.8				93.8	3.8	Tulonen and Vuorinen (1996)
Welsh rivers (UK)	1993		10.0				6.0	6.0	22.8			Weatherley et al. (1997)

Tnona refers to transnonachlor.
[a] Value represents average of the data.
[b] Dry weight basis.

Table 5
Reported concentrations of heavy metals in *Anguilla anguilla* in Europe

Sampling site	Year	Mercury	Cadmium	Lead	Copper	Zinc	Arsenic	Reference
River Turia (Spain)	2000		4.9	101.8	977	16,950	227.9	Bordajandi et al. (2003)
River Gironde (France)	2001	170			150	10,200		Durrieu et al. (2005)
Mersey estuary (UK)	1991–1993	962			884	24,000	1100	Collings et al. (1996)[a]
River Ferrerias and River Raices (Spain)	No date given	278	24.5	33.25	218.75			Linde et al. (2004)[a]

[a] Value represents average of the data.

De Boer et al. (1993) demonstrated a highly significant, empirical relationship between the concentration of PCB congener 153 in ng g^{-1} wet weight and pg PCB TEQ g^{-1} fresh weight. Here, we used their equation to assess the risk of eel consumption. In this study, eel showed an average PCB 153 concentration of 223 ng g^{-1} wet weight, which corresponds to 17.1 pg PCB TEQ g^{-1} wet weight. So even without accounting for the presence of dioxins and furans, an average sample of muscle meat of eel captured in surface waters exceeds the maximum by a factor of two.

A meal consisting of 100 g would result in a dietary uptake of 24 pg TEQ g^{-1} body weight for an adult person weighing 70 kg. It follows that dietary exposure to PCBs by eating wild eel caught by angling exceeds the tolerable weekly intake that was advanced by the Scientific Committee on Food of the EU, which is 14 pg TEQ kg^{-1} body weight (Communication/593/2001). These results do not take into account the average dietary intake of dioxins and dioxin-like PCBs in the EU which was in the range of 1.2–3 pg kg^{-1} body weight per day. From that perspective, the consumption of eel caught in the wild should continue to be discouraged.

Table 6
Maximum residue and contaminant levels (MRL) in eel as adopted by different European regulations and proportion of eel (%) captured in study with tissue concentrations higher than this maximum

Substance	MRL in eel or fish	Proportion of non-compliant eel (%)
Pesticides (EEC/86/1986)		
Endrin	0.01 mg kg^{-1} wet weight	3.8
Dieldrin	0.2 mg kg^{-1} wet weight	0.4
α-HCH	0.02 mg kg^{-1} wet weight	0
Lindane	0.2 mg kg^{-1} wet weight	2.3
Sum of DDTs	1 mg kg^{-1} wet weight	0.5
HCB	0.1 mg kg^{-1} wet weight	0.1
Transnonachlor	0.05 mg kg^{-1} wet weight	0
Heavy metals (EC/466/2001)		
Mercury	1 mg kg^{-1} wet weight	0
Lead	0.4 mg kg^{-1} wet weight	2.4
Cadmium	0.1 mg kg^{-1} wet weight	1.0
Dioxines and PCBs (EC/199/2006)		
Sum of dioxins	4 pg WHO-TEQ g^{-1} wet weight	–
Sum of dioxines and dioxin-like PCBs	12 pg WHO-TEQ g^{-1} wet weight	32.0[a]
Sum of seven indicator PCBs	0.075 mg kg^{-1} wet weight[b]	75.4

[a] Based on a regression equation between PCB 153 and PCB TEQ (see text).
[b] Belgian MRL.

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